

EFFECT OF HEAT TREATMENTS ON SILICON DIOXIDE AND SILICON NITRIDE NANOCOMPOSITES

MUHAMMAD ISAMUDDIN BIN AHMAD RITZZNEY

A project report submitted in partial fulfilment of the
requirements for the award of the degree of
Master of Engineering (Electrical Power)

School of Electrical Engineering
Faculty of Engineering
Universiti Teknologi Malaysia

JANUARY 2019

DEDICATION

This project report is specially dedicated to my beloved father and mother who always believe in me, love, and gave utmost support to finish my study. Also, to my fiancée who always love, advice and pushed me till the end through this long run of the study to the finish line. Not to forget, to my brothers, sisters and colleagues who share the same passion and gave moral support among our self to finish this great journey in UTM

ACKNOWLEDGEMENT

In preparing this project report, I was in contact with many people, researchers, and practitioners. They have contributed towards my understanding and thoughts. In particular, I wish to express my sincere appreciation to my project report supervisor, Ir. Dr. Lau Kwan Yiew, for encouragement, guidance, critics and friendship. Without his continued support and interest, this project report would not have been the same as presented here.

My fellow postgraduate student should also be recognised for their support. My sincere appreciation also extends to all my colleagues and others who have provided assistance at various occasions. Their views and tips are useful indeed. Unfortunately, it is not possible to list all of them in this limited space.

Last but not least, I would like to thank to my beloved family and fiancée who always have been supporting me to the end. Word are powerless to express my gratitude towards every one of you.

ABSTRACT

This project determined the effect of breakdown strength when heat treatment process was applied on nanocomposites. Unlike the normal approach where nanocomposites rarely takes into account the effect of heat treatment process, the heat treatment method used in this project was by heating to nanocomposites under 3 conditions of temperature. The proposed approach utilized nanosilica and nanosilicon nitride as the nanofillers and polyethylene blend as the base polymer with the nanocomposites exposed to heat treatment under vacuum at temperatures of 80°C, 90°C and 100°C. The results showed that unfilled polyethylene blend had the highest measurement of breakdown strength which were 27.82% and 10.89% higher when compared with the addition on 1 wt% nanosilica and 1 wt% nanosilicon nitride at ambient temperature. However, when the thin film samples were treated with 3 types of heat treatment in vacuum condition and underwent DC breakdown test afterwards, the results for samples treated at 90°C improved, where the addition of 1 wt% nanosilica and 1 wt% nanosilicon nitride increased to 3.45% and 28.88% when compared with unfilled polyethylene blend. This meant the conditioning of heat treatment could improve the characteristic of the dielectric properties of the nanocomposites. The work can be considered significant due to the fact that the breakdown strength of nanocomposites will be affected by the type of nanofillers used and heat treatment process on the nanocomposites.

ABSTRAK

Projek ini menentukan kesan proses rawatan haba kepada kekuatan pecah tebat komposit nano. Tidak seperti pendekatan biasa di mana komposit nano jarang mengambil kira kesan proses rawatan haba, kaedah rawatan haba yang digunakan di dalam projek ini ialah dengan memanaskan komposit nano di bawah 3 keadaan suhu. Pendekatan yang dicadangkan adalah dengan menggunakan silika nano dan silikon nitrida nano sebagai pengisi nano dan polietilena sebagai polimer asas dengan komposit nano terdedah kepada rawatan haba di bawah vakum pada 80, 90 dan 100 darjah suhu. Keputusan menunjukkan bahawa campuran polietilena yang tidak terisi mempunyai kekuatan pecah tebat tertinggi iaitu 27.82% dan 10.89% lebih tinggi berbanding dengan penambahan pada 1 wt% silika nano dan 1 wt% silikon nitrida nano pada suhu keadaan semasa. Walau bagaimanapun, apabila spesimen filem dirawat dengan 3 jenis rawatan haba dalam keadaan vakum dan menjalani ujian pecah tebat arus terus, keputusan untuk spesimen yang dirawat pada 90 darjah suhu menunjukkan peningkatan, di mana penambahan 1 wt% silika nano dan 1 wt% silikon nitride nano meningkat kepada 3.45% dan 28.88% apabila dibandingkan dengan campuran polietilena tidak terisi. Ini bermakna penyelenggaraan rawatan haba dapat meningkatkan ciri sifat penebat elektrik komposit nano. Kerja ini boleh dianggap penting kerana kekuatan pecahan komposit nano akan dipengaruhi dengan jenis pengisi nano yang digunakan serta proses rawatan haba yang didedahkan kepada komposit nano.

TABLE OF CONTENTS

	TITLE	PAGE
	DECLARATION	ii
	DEDICATION	iii
	ACKNOWLEDGEMENT	iv
	ABSTRACT	v
	ABSTRAK	vi
	TABLE OF CONTENT	vii
	LIST OF TABLES	x
	LIST OF FIGURES	xi
	LIST OF ABBREVIATIONS	xiii
	LIST OF SYMBOLS	xiv
CHAPTER	1 INTRODUCTION	1
	1.1 Background of Project	1
	1.2 Problem Statement	2
	1.3 Project Objectives	3
	1.4 Scope of Project	3
	1.5 Gantt's Chart for Project 1 and 2	4
	1.6 Proposal Outline	5
CHAPTER	2 LITERATURE REVIEW	7
	2.1 Introduction	7
	2.2 Electrical Breakdown of Insulation	8
	2.3 Properties of Polymer Nanocomposites	9
	2.4 Polymer Nanocomposites as Insulation	10
	2.5 Processing Method	12
	2.6 Silica Dioxide Nanofiller	12
	2.7 Silicon Nitride Nanofiller	13

	2.8	The Complexity of Polymer Nanocomposites Materials	13
	2.9	Heat Treatment on Polymer Nanocomposites	15
	2.10	Weibull Parameter	16
CHAPTER	3	RESEARCH METHODOLOGY	17
	3.1	Introduction	17
	3.2	Process Flow for Formulation of Nanocomposites	17
	3.3	Material Preparation	20
	3.4	Equipment For Fabrication Process	21
	3.4.1	Weighting Scale Machine	21
	3.4.2	Two-roll Mill Machine	22
	3.4.3	Hydraulic Press Machine	23
	3.5	Equipment for Heat Treatment under Vacuum	24
	3.6	DC Breakdown Test Configuration and Equipment	24
	3.6.1	AC/DC HV Test Set	25
	3.6.2	Tesla Coil Step up Transformer	26
	3.6.3	Spherical Electrodes	26
	3.6.4	Transformer Oil	27
CHAPTER	4	RESULT AND DISCUSSION	29
	4.1	Introduction	29
	4.2	The Effect of the Heat Treatment on the Mass of the Nanocomposites	29
	4.3	DC Breakdown Strength of Nanocomposites	32
	4.3.1	Nanocomposites under Ambient Temperature	32

	4.3.2	Nanocomposites with Heat Treatment under Vacuum - 80°C	34
	4.3.3	Nanocomposites with Heat Treatment under Vacuum - 90°C	36
	4.3.4	Nanocomposites with Heat Treatment under Vacuum - 100°C	38
	4.4	Performance of Nanocomposites in DC Breakdown Strength	40
CHAPTER	5	CONCLUSION	43
	5.1	Conclusion	43
	5.2	Future Recommendation	44
REFERENCES			45

LIST OF TABLES

TABLE NO.	TITLE	PAGE
Table 1.1	Project Schedule for Project 1	4
Table 1.2	Project Schedule for Project 2	4
Table 2.1	Comparison Between Nanocomposites and Microcomposites	9
Table 3.1	Nanocomposites Sample Weightage	21
Table 4.1	The Effect of the Heat Treatment on the Mass of the Nanocomposites	30
Table 4.2	The Weibull Distribution Parameter of Ambient Temperature Condition	32
Table 4.3	The Weibull distribution parameter of heat treatment (80°C) in vacuum condition	34
Table 4.4	The Weibull distribution parameter of heat treatment (90°C) in vacuum condition	36
Table 4.5	The Weibull distribution parameter of heat treatment (100°C) in vacuum condition	38
Table 4.6	DC breakdown strength of all nanocomposites in different types of conditioning	40
Table 4.7	Surrounding ambient temperature during DC breakdown test measurement	42

LIST OF FIGURES

FIGURE NO.	TITLE	PAGE
Figure 2.1	Polyethylene Chain	11
Figure 3.1	Process Flow for Nanocomposites Samples Preparation	19
Figure 3.2	(a) LDPE (b) HDPE (c) Nanosilica Powder (d) Nanosilicon Nitride Powder	20
Figure 3.3	Weighting Scale Machine	22
Figure 3.4	Two-roll Mill Machine	23
Figure 3.5	Hydraulic Press Machine	23
Figure 3.6	Constance Heat Vacuum Oven	24
Figure 3.7	The Breakdown Test Configuration	25
Figure 3.8	AC/DC HV Test Set	25
Figure 3.9	Tesla Coil Step Up Transformer	26
Figure 3.10	Spherical Electrode with Holding Mechanism	27
Figure 3.11	The Film Samples Immersed in Transformer Oil	27
Figure 4.1	The Change of Mass (%) of All Nanocomposites Under Heat Treatment in Vacuum Condition	31
Figure 4.2	The Weibull Distribution of the DC Breakdown Strength of Nanocomposites for Ambient Temperature Condition	33
Figure 4.3	The Weibull distribution of the DC Breakdown Strength of Nanocomposites Under Heat Treatment (80°C) in Vacuum Condition	35
Figure 4.4	The Weibull distribution of the DC Breakdown Strength of Nanocomposites Under Heat Treatment (90°C) in Vacuum Condition	37

Figure 4.5	The Weibull distribution of the DC Breakdown Strength of Nanocomposites Under Heat Treatment (100°C) in Vacuum Condition	39
Figure 4.6	Nanocomposites DC Breakdown Strength	42

LIST OF ABBREVIATIONS

SiO ₂	-	Silica
Si ₃ N ₄	-	Silicon Nitride
PE	-	Polyethylene
LDPE	-	Low Density Polyethylene
HDPE	-	High Density Polyethylene
AC	-	Alternating Current
DC	-	Direct Current
HV	-	High Voltage

LIST OF SYMBOLS

wt%	-	Weightage
°C	-	Degree Celsius
kV	-	Kilovoltage
g	-	Gram
exp	-	Exponential
α	-	The Breakdown Strength of the Distribution
β	-	The Best Repeatability or Shape Parameter
E	-	Dielectric Strength
P (x)	-	Failure Accumulated Probability
m	-	Metre
mm	-	Millimetre
μm	-	Micrometre

CHAPTER 1

INTRODUCTION

1.1 Background of Project

A composite mainly comprises a polymer that yields great mechanical and electrical properties. In high voltage applications, conventional composites are ideal to be used for constructing dielectric materials and high voltage insulation. However, in long term applications, conventional composites are often prone to corrosion, tracking, degradation due to heat radiation, and low resistance against surface pollution and acidic open air environment. Therefore, research needs to be carried out to overcome these problems.

A nanocomposite is a polymer based material which have been added with nano-size particles. It is also known as a nanodielectric, which was first introduced by Lewis in 1994 [1]. This material is regarded as a great breakthrough in the scholastic community as it possesses superior mechanical and electrical properties in comparison with conventional composite materials. Besides that, it has a higher breakdown strength, higher permittivity potential depending on the types of nano-fillers used, and better resistance against surface discharge effect. However, the performance of nanodielectric under long term use subjected to multiple stresses such as heat temperature, ultra violet radiation, electrical stress, mechanical stress, and moisture in open environment, have not been thoroughly investigated.

In relation to this, numerous experiments have focused on investigating the dielectric behaviours of this material system. Despite numerous positive experimental results reported on the use of nanocomposites as electrical insulating materials, there exists many fundamental challenges remain to be addressed. In this study, the effects of various heat processes on nanocomposites containing different types of oxide-based nano-fillers were investigated.

1.2 Problem Statement

It is known that the absorption of water on the nanocomposites can reduce its breakdown strength. By applying treatments such as heat treatment, their breakdown strength performance may be improved. However, the performance of nanocomposites subjected to heat treatment were less investigated in existing works. Other than that, there likely exists different breakdown strengths for nanocomposites when they are exposed to different heat temperatures. Related to this, the effect of heat treatment on the silicon-based nano-fillers on the nanocomposite needs to be thoroughly investigated. This is because the outcome might differ since the breakdown strength of nanocomposites is influenced by the treatment and types of nano-fillers used. Besides that, there are also fundamental challenges yet to be addressed in this field.

1.3 Project Objectives

The objectives of this project are as follows:-

- i. To formulate unfilled polyethylene blend, nanosilica filled polyethylene blend and nanosilicon nitride filled polyethylene blend.
- ii. To determine breakdown strength between unfilled polyethylene blend with nanosilica filled polyethylene blend and nanosilicon nitride filled polyethylene blend.
- iii. To examine the effect of heat treatment on breakdown strength of unfilled polyethylene blend, nanosilica filled polyethylene blend and nanosilicon nitride filled polyethylene blend.

1.4 Scope of Project

The study utilized nanosilica (SiO_2) and nanosilicon nitride (Si_3N_4) as an instance of silicon based nano-filler and polyethylene blend (PE blend) as an instance of polymer which consisted of 80% low density polyethylene (LDPE) with 20% high density polyethylene (HDPE). The weightages of the silicon based nano-filler used were 1 wt%. The heat treatment process on the nanocomposites samples was done in vacuum oven at 80°C, 90°C and 100°C in a duration of 16 hours. The breakdown strength test was conducted in DC system and the increment of the voltage was 2 kV for every 20 seconds until the sample failed.

1.5 Gantt's Chart for Project 1 and 2

The project 1 and project 2 schedule can be refer to the Table 1.1 and Table 1.2 below.

Table 1.1: Project schedule for project 1

No	Activity	Week														
		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
1	Project title and supervisor selection															
2	List for project title and supervisor released															
3	Brainstorm on project title															
4	Decide scope of project															
5	Synopsis submission															
6	Prepare literature review															
7	Prepare project methodology															
8	Preparation of sample															
9	Project proposal submission															
10	Seminar presentation															
11	Project report submission															

Table 1.2: Project schedule for project 2

No	Activity	Week														
		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
1	Understand the experiment and safety procedure															
2	Conduct breakdown test															
3	Interpreting and analyse data															
4	Arrange and conclude the results															
5	Prepare for seminar															
6	Seminar presentation															
7	Thesis preparation															

1.6 Proposal Outline

This report contains five chapters. Chapter 1 is known as an introduction to the research project which consists of 5 subtopics. The subtopic covers background of project, problem statements, project objectives, scopes and Gantt's charts.

As for chapter 2, it is called literature review. Literature review is the study about the background of project based on past research studies concerning on the fundamental and issues on nanocomposites. This chapter covers 10 subtopics. The subtopics consist of introduction, electrical breakdown of insulation, properties of polymer nanocomposites, polymer nanocomposites as insulation, processing method, silica dioxide nanofiller, silicon nitride nanofiller, the complexity of polymer nanocomposites material, heat treatment on polymer nanocomposites, and Weibull analysis.

Chapter 3 is the research methodology which provides in details on how to accomplish the objectives of this study. It consists of 6 subtopics which covers on introduction, process flow for formulation of nanocomposites, material preparation, equipment for fabrication process, equipment for heat treatment, and DC breakdown test configuration and equipment

Next, chapter 4 is known as result and discussion which conveys the result based on preparation method on chapter 3. This chapter consists of 4 subtopics of introduction, the effect of heat treatment on the mass of the nanocomposites, DC breakdown strength nanocomposites, and performance of nanocomposites in DC breakdown strength.

Lastly, for chapter 5. Chapter 5 is the overall conclusion for the project 1 and 2. It is the wrap up of all works from chapter 1 to 4 based on past research studies to attain this project objectives successfully. Also, it include future recommendation as to improve the present research works.

REFERENCES

- [1] T. Tanaka, "Dielectric nanocomposites with insulating properties," *IEEE Transactions on Dielectrics and Electrical Insulation*, vol. 12, no. 5, pp. 914-928, 2005.
- [2] M. S. Naidu, *High voltage engineering*: Tata McGraw-Hill Education, 2013.
- [3] I. Z. Intan, "Breakdown characteristics of low density polyethylene nanocomposites containing different sizes of nanosilica," University Technology Malaysia, 2017.
- [4] T. Tanaka, Y. Matsuo, and K. Uchida, "Partial discharge endurance of epoxy/SiC nanocomposite." pp. 13-16.
- [5] T. Tanaka, G. Montanari, and R. Mulhaupt, "Polymer nanocomposites as dielectrics and electrical insulation-perspectives for processing technologies, material characterization and future applications," *IEEE transactions on Dielectrics and Electrical Insulation*, vol. 11, no. 5, pp. 763-784, 2004.
- [6] M. Roy, J. Nelson, R. MacCrone, L. S. Schadler, C. Reed, and R. Keefe, "Polymer nanocomposite dielectrics-the role of the interface," *IEEE transactions on dielectrics and electrical insulation*, vol. 12, no. 4, pp. 629-643, 2005.
- [7] P. Dittanet, and R. A. Pearson, "Effect of silica nanoparticle size on toughening mechanisms of filled epoxy," *Polymer*, vol. 53, no. 9, pp. 1890-1905, 2012.
- [8] C. Zhang, R. Mason, and G. C. Stevens, "Dielectric properties of epoxy and polyethylene nanocomposites." pp. 393-396.
- [9] P. M. Ajayan, L. S. Schadler, and P. V. Braun, *Nanocomposite science and technology*: John Wiley & Sons, 2006.
- [10] K. Lau, A. Vaughan, and G. Chen, "Nanodielectrics: opportunities and challenges," *IEEE Electrical Insulation Magazine*, vol. 31, no. 4, pp. 45-54, 2015.
- [11] R. Smith, C. Liang, M. Landry, J. Nelson, and L. Schadler, "The mechanisms leading to the useful electrical properties of polymer nanodielectrics," *IEEE Transactions on Dielectrics and Electrical Insulation*, vol. 15, no. 1, 2008.
- [12] K. Y. Lau, "Structure and electrical properties of silica-based polyethylene nanocomposites," University of Southampton, 2013.
- [13] I. L. Hosier, "Morphology and electrical properties of polyethylene blends," University of Reading, 1996.

- [14] K. A. Narh, A.-T. Agwedicham, L. J. Jallo, K. Y. Rhee, and J. H. Lee, "Influence of deagglomeration states of carbon nanotubes on the thermal and mechanical properties of carbon nanotube-reinforced polyethylene oxide," *International Journal of Nanoscience*, vol. 8, no. 01n02, pp. 23-27, 2009.
- [15] G. Xu, J. Wang, X. Ji, J. Xiong, and F. Li, "Effect of nano-silicon nitride on the mechanical and electric properties of polypropylene nanocomposite," *Journal of composite materials*, vol. 41, no. 18, pp. 2213-2223, 2007.
- [16] K. Lau, A. Vaughan, G. Chen, I. Hosier, and A. Holt, "On the dielectric response of silica-based polyethylene nanocomposites," *Journal of Physics D: Applied Physics*, vol. 46, no. 9, pp. 095303, 2013.
- [17] I. Hosier, M. Praeger, A. Vaughan, and S. Swingler, "The effects of hydration on the DC breakdown strength of polyethylene composites employing oxide and nitride fillers," *IEEE Transactions on Dielectrics and Electrical Insulation*, vol. 24, no. 5, pp. 3073-3082, 2017.
- [18] W. Wang, S. Li, F. Tang, and J. Li, "Characteristics on breakdown performance of polyethylene/silica dioxide nanocomposites." pp. 521-524.
- [19] H. Alamri, and I. M. Low, "Effect of water absorption on the mechanical properties of nano-filler reinforced epoxy nanocomposites," *Materials & Design*, vol. 42, pp. 214-222, 2012.
- [20] K. Lau, A. Vaughan, G. Chen, and I. Hosier, "Dielectric response of polyethylene nanocomposites: The effect of surface treatment and water absorption." pp. 275-278.
- [21] S. Raider, R. Flitsch, J. Aboaf, and W. Pliskin, "Surface oxidation of silicon nitride films," *Journal of The Electrochemical Society*, vol. 123, no. 4, pp. 560-565, 1976.
- [22] W. Zhou, C. Wang, T. Ai, K. Wu, F. Zhao, and H. Gu, "A novel fiber-reinforced polyethylene composite with added silicon nitride particles for enhanced thermal conductivity," *Composites Part A: Applied Science and Manufacturing*, vol. 40, no. 6-7, pp. 830-836, 2009.
- [23] I. L. Hosier, M. Praeger, A. S. Vaughan, and S. G. Swingler, "The effects of water on the dielectric properties of silicon-based nanocomposites," *IEEE Transactions on Nanotechnology*, vol. 16, no. 2, pp. 169-179, 2017.
- [24] M. Praeger, I. Hosier, A. Vaughan, and S. Swingler, "The effects of surface hydroxyl groups in polyethylene-silica nanocomposites." pp. 201-204.
- [25] T. Tanaka, A. Bulinski, J. Castellon, M. Frechette, S. Gubanski, J. Kindersberger, G. C. Montanari, M. Nagao, P. Morshuis, and Y. Tanaka, "Dielectric properties of XLPE/SiO₂ nanocomposites based on CIGRE WG D1. 24 cooperative test results," *IEEE Transactions on Dielectrics and Electrical Insulation*, vol. 18, no. 5, pp. 1482-1517, 2011.

- [26] R. Abernethy, "The New Weibull Handbook (1996)," *ISBN 0-965*, 1966.
- [27] I. Hosier, M. Praeger, A. Vaughan, and S. Swinger, "Electrical properties of polymer nano-composites based on oxide and nitride fillers." pp. 438-441.
- [28] M. A. N. Zahid, "Electrical Breakdown Performance Of Polyethylene Blends With Different Nanofillers," Universiti Teknologi Malaysia, 2017.
- [29] K. Lau, M. Piah, and K. Ching, "Correlating the breakdown strength with electric field analysis for polyethylene/silica nanocomposites," *Journal of Electrostatics*, vol. 86, pp. 1-11, 2017.
- [30] C. Huang, and Q. Cheng, "Learning from nacre: constructing polymer nanocomposites," *Composites Science and Technology*, vol. 150, pp. 141-166, 2017.
- [31] J. K. Nelson, *Dielectric polymer nanocomposites*: Springer, 2010.